

Scattering laser light on cold atoms: multiple scattering signals from single-atom responses

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We deduce the coherent backscattering signal from two distant laser-driven atoms using single-atom equations. In contrast to the standard master equation treatment, this new approach is suitable for the generalization to a large number of atomic scatterers.

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Cooled atomic gases have been successfully used in experimental studies of coherent backscattering (CBS) of laser light [1], and are considered to be a promising medium for the observation of Anderson localization [2] with photons. Apart from their fundamental interest, these interference phenomena in disordered systems are important for practical applications such as random lasers [3] and quantum memory for light [4]. Inasmuch as *multiple* scattering of photons is crucial for the emergence of weak and strong localization [5] in disordered atomic clouds, while it can play a detrimental role for the storage of quantum information [6], an appropriate theory taking into account all relevant effects of atom-light interactions is in order.

The theory of multiple scattering in dilute media that consist of a disordered collection of discrete scatterers relies on the division of the total scattering process into single scattering events. These are described by a scattering operator which is usually assumed to be identical for all the individual scatterers, and thereby serves as the fundamental building block for the total multiple scattering process [7]. This approach, however, fails in the case of atomic scatterers, for the following two reasons: first, due to the saturation of the atomic transition with increasing intensity of the incident light, the atoms scatter light nonlinearly. The outgoing field (E_{out}) is not proportional to the incoming one (E_{in}), and therefore cannot be described by a linear scattering operator of the form $E_{\text{out}} = TE_{\text{in}}$. Second, the light scattered by near-resonant atoms exhibits fluctuations due to the quantum mechanical coupling of the atoms to the electromagnetic vacuum. This means that even if the incident laser light E_{in} is perfectly coherent, this is not the case for the scattered field E_{out} . In particular, its average intensity differs from the square of the average field, i.e., $\langle |E_{\text{out}}|^2 \rangle > |\langle E_{\text{out}} \rangle|^2$. The difference defines the incoherent (or inelastic) component of the resonance fluorescence intensity.

So far, no satisfying theory exists for incorporating both these effects into a multiple scattering approach. Recently [8, 9], a theory for coherent backscattering by nonlinear, classical scatterers was presented, but does not take into account any quantum fluctuations due to in-

elastic scattering. A perturbative approach based on the scattering matrix of two photons was proposed in [10], but is only valid if incident light intensity and optical thickness of the atomic medium are small. On the other hand, standard tools of quantum optics (master equations, optical Bloch equations ...) are well adapted to describe the atom-field interaction for arbitrary intensities of the incident field, but are restricted to a small number of atoms coupled to each other by photon exchange [11, 12]. This is due to the fact that the dimension of the atomic Hilbert space grows exponentially with the number of atoms.

In the present Letter, we show that - for the case of two randomly placed atoms with mean distance much larger than the laser wavelength - the two perspectives of nonlinear multiple scattering and open system dynamics can be unified within a single approach. Our results agree perfectly well with the master equation and quantum Langevin treatments presented in [13, 14]. In contrast to [13, 14], however, the present approach can be generalized for a large number of atoms.

Imagine a cloud of atoms in free space which is excited by a coherent laser beam. We are interested in the average intensity radiated into different directions θ , where the average is taken over the random positions of the atoms. Around the backscattering direction $\theta = 0$, a coherent backscattering (CBS) peak is observed [1], originating from the constructive interference of counterpropagating, multiply scattered waves. In the case of a dilute and optically thin cloud, the leading contribution to the interference signal comes from double scattering events, where the laser interacts only with pairs of atoms. (Single scattering does not contribute to CBS, since direct and reversed paths are identical in this case.) Hereafter, we therefore derive the double scattering CBS contribution from a simplified model composed of only two atoms, considered stationary for the time for one scattering cycle to take place, and separated by a distance which is large compared to the laser wavelength [11, 12]. For simplicity, we further consider two-level atoms coupled to a scalar photon field, such as to avoid the additional (though straightforward) technical overhead needed to account for photon polarization and degenerate atomic

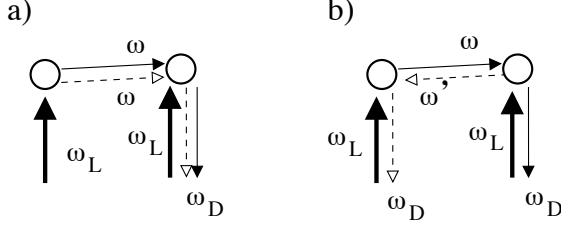


Figure 1: Double scattering contributions to coherent backscattering. The thick arrows represent the intense laser beam, and the thin solid and dashed arrows the Fourier components of the scattered fields, $E^{(+)}(t)$ and $E^{(-)}(t)$, respectively. (a) One of the atoms emits a photon with frequency ω which is scattered by the other atom before it reaches the detector. This process yields the background intensity $I_L(\omega_D)$ of the photodetection signal. (b) One of the atoms emits a photon with frequency ω , the other a photon with frequency $\omega' = \omega_L + \omega_D - \omega$ (see text). This process gives rise to the coherent backscattering interference peak $I_C(\omega_D)$.

transitions.

The situation we consider is summarized in Fig. 1. Two atoms scatter photons injected through an intense laser beam at frequency ω_L into distinct modes with frequencies ω , ω' , and ω_D . While ω_D is the frequency of the detected photons, ω and ω' are the frequencies of the (unobserved) photons exchanged between the atoms – thus mediating the double scattering process. Fig. 1a defines the weakly angle-dependent background of the photodetection intensity, also called “ladder intensity” I_L in the following, whereas Fig. 1b determines the height I_C (“crossed intensity”) of the coherent backscattering peak around $\theta = 0$, resulting from interference between photons travelling along reversed scattering paths. Both I_L and I_C are independent of the positions of the two atoms, and thus define, for the case of large mean distance between the atoms, the configuration-averaged photodetection intensity [11, 12]. The assumption of large distance is required in order to neglect processes where more than one photon is exchanged between the atoms.

The key ingredient for our subsequent treatment is the observation that the detected double scattering signal can be deduced from the superposition of the fields scattered by both individual atoms into the detector, each atom being subject to a *bichromatic* driving field $\mathcal{E}(t)$ composed of the injected laser *and* the photons scattered by the other atom,

$$\mathcal{E}(t) = \mathcal{E}_L e^{-i\omega_L t} + v^{(+)} e^{-i\omega t} + \mathcal{E}_L^* e^{i\omega_L t} + v^{(-)} e^{i\omega t}. \quad (1)$$

The classical field $\mathcal{E}(t)$ is decomposed into its positive and negative frequency components with complex amplitudes \mathcal{E}_L and $v^{(+)} (= v^{(-)*})$, since the final figure of merit, the average stationary intensity $\langle I \rangle = \lim_{t \rightarrow \infty} \langle E^{(-)}(t) E^{(+)}(t) \rangle$ measured by the detector, is conveniently expressed in terms of positive and negative frequency components of the signal radiated by the two atoms.

The single atom response to the bichromatic drive is characterized by the associated cross correlation function in frequency space, $C(\omega_1, \omega_2) = \langle \tilde{E}^{(-)}(\omega_1) \tilde{E}^{(+)}(\omega_2) \rangle$, which correlates the, in general, different frequencies of the negative (thin dashed lines in Fig. 1) and positive (thin continuous lines in Fig. 1) frequency components $\tilde{E}^{(-)}(\omega_1)$ and $\tilde{E}^{(+)}(\omega_2)$ of the emitted field (left atom: $\omega_1 = \omega_D$ and $\omega_2 = \omega$, right atom: $\omega_1 = \omega'$ and $\omega_2 = \omega_D$, in Fig. 1b). Since the emitted fields $E^{(-)}$ and $E^{(+)}$, in turn, are proportional to the atomic dipole raising and lowering operators σ_+ and σ_- , the frequency correlation function can be written as:

$$C(\omega_1, \omega_2) = \int_{-\infty}^{\infty} \frac{dt_1 dt_2}{2\pi} e^{-i\omega_1 t_1 + i\omega_2 t_2} \langle \sigma_+(t_1) \sigma_-(t_2) \rangle, \quad (2)$$

and is thus given by the solutions $\sigma_+(t)$ and $\sigma_-(t)$ of the *single atom* optical Bloch equations under the bichromatic drive, Eq. (1).

The total detected double scattering intensity at frequency ω_D is finally given by an integral over the product of the cross correlation functions of both atoms, where the integral runs over the frequencies of the exchanged photons. For an explicit evaluation of these integrals, we expand $C(\omega_1, \omega_2)$ to lowest order in $v^{(+)}$ and $v^{(-)}$. This perturbative treatment is actually already implied by our ansatz of *classical* scattered fields in Eq. (1), which neglects their quantum statistical properties – encoded in the second order correlation functions, i.e. in higher orders of $v^{(\pm)}$ – and is certainly justified in the presently considered limit of a dilute gas with large interatomic distances and, thus, $|v^{(\pm)}| \ll |\mathcal{E}_L|$. Solving Eq. (2) for the case of bichromatic driving, Eq. (1), and keeping only terms up to first order in $v^{(+)}$ and/or $v^{(-)}$, we obtain the following four terms:

$$C|_{v^{(\pm)}=0} = \delta(\omega_1 - \omega_2) P_0(\omega_1), \quad (3a)$$

$$\left. \frac{\partial C}{\partial v^{(+)}} \right|_{v^{(\pm)}=0} = \delta(\omega_2 - \omega_1 - \omega + \omega_L) P_1(\omega; \omega_2), \quad (3b)$$

$$\left. \frac{\partial C}{\partial v^{(-)}} \right|_{v^{(\pm)}=0} = \delta(\omega_1 - \omega_2 - \omega + \omega_L) P_1^*(\omega; \omega_1), \quad (3c)$$

$$\left. \frac{\partial^2 C}{\partial v^{(+)} \partial v^{(-)}} \right|_{v^{(\pm)}=0} = \delta(\omega_1 - \omega_2) P_2(\omega; \omega_1). \quad (3d)$$

The δ -functions in Eqs. (3a-3d) originate from integrating over $t_+ := t_1 + t_2$ in Eq. (2), and are thus a consequence of time translation invariance or energy conservation. They ensure that the negative and positive frequency components of the field are radiated at the same frequency ω in Fig. 1a, whereas $\omega' = \omega_L + \omega_D - \omega$ in Fig. 1b.

$P_0(\omega)$ in Eq. (3a) denotes the usual resonance fluorescence spectrum for monochromatic driving including both the elastic and inelastic components [16], and is associated with the emission of a fluorescence photon of frequency ω by the left atom in Fig. 1a. $P_2(\omega; \omega_D)$, defined by Eq. (3d), describes the subsequent scattering cross section of this fluorescence photon by the right atom, into the finally detected frequency mode ω_D . Consequently,

the final expression for the ladder contribution (Fig. 1a) to the total scattered intensity at frequency ω_D reads

$$I_L(\omega_D) = |g|^2 \int d\omega P_0(\omega) P_2(\omega; \omega_D), \quad (4)$$

where $g = \exp(i\omega_L r)/(k_L r)$, with $k_L = \omega_L/c$, describes the effective coupling strength between the two atoms. The frequency dependence of g can be neglected for inverse propagation times c/r between the two atoms much larger than the frequency differences $\omega' - \omega$ between the exchanged photons (which is the case for typical experimental parameters). Note that the integral over the frequency ω of the photon emitted by the left atom expresses the correlation between both atoms induced through the photon exchange.

Analogously, according to Eq. (3c), $P_1^*(\omega'; \omega_D)$ represents the emission of a negative frequency *amplitude* ω_D (incident on the detector) and a positive frequency *amplitude* ω (towards the other atom) by the left atom in Fig. 1b, which, in turn, is subject to a negative frequency amplitude ω' originating from the right atom. The right atom, subject to a positive frequency amplitude ω from the left atom, radiates a negative frequency amplitude ω' (towards the left atom) and a positive frequency amplitude ω_D (into the detector), represented by $P_1(\omega; \omega_D)$ in Eq. (3b). Energy conservation further enforces $\omega' = \omega_L + \omega_D - \omega$. For the crossed contribution to the total scattered intensity at the frequency ω_D , this entails

$$I_C(\omega_D) = |g|^2 \int d\omega P_1^*(\omega_L + \omega_D - \omega; \omega_D) P_1(\omega; \omega_D). \quad (5)$$

Once again, both atomic dipoles are conditioned on each other through the frequency of the exchanged photon. Finally, the total intensities measured by a broadband detector are obtained by integrating over the detected photon frequency, i.e., $I_{L,C} = \int d\omega_D I_{L,C}(\omega_D)$.

By numerical integration of Eqs. (4) and (5), we have studied the behavior of the elastic and inelastic components of the ladder and crossed terms for different values of the laser-atom detuning $\delta = \omega_L - \omega_0$ (with ω_0 the atomic resonance frequency) and of the Rabi frequency Ω (which is proportional to the laser amplitude $|\mathcal{E}_L|$). In particular, some examples of the normalized *inelastic* ladder and crossed spectra for several parameters of the driving field are presented in Fig. 2. At small Rabi frequencies (see Fig. 2(a)), the background and interference spectra are qualitatively very similar to the analogous quantities in the helicity preserving polarization channel [10, 13], and can be deduced from the two-photon scattering amplitudes. When $\Omega \gg \gamma$ (Fig. 2(b)), the spectral features of the ladder and crossed terms can be interpreted in terms of scattering and self-interference of photons emitted by one atom on the dressed state levels of the other atom [13]. Note that negative values of the ladder term in certain spectral regions arises from absorption dominating emission for doubly scattered photons; the total contribution, including single scattering, is always positive.

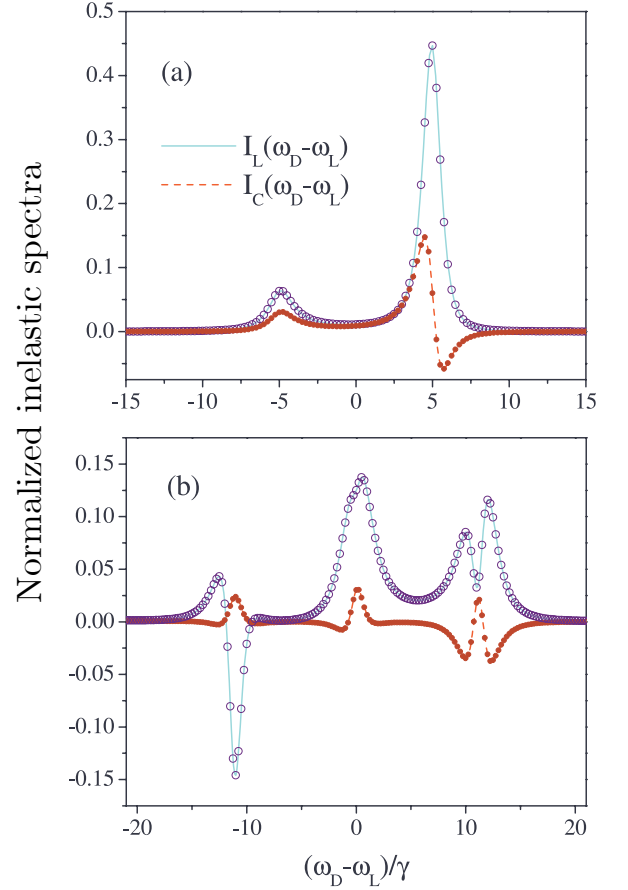


Figure 2: (Color online) Inelastic double scattering spectra as obtained from the two-atom master equation (solid line: background, dashed line: interference contribution), compared to the predictions of our new approach, Eqs. (4,5), based on single-atom Bloch equations with bichromatic driving (open circles: background, filled circles: interference contribution). Laser-atom detuning $\delta = -5\gamma$, Rabi frequency $\Omega =$ (a) 0.1γ and (b) 10γ . Perfect agreement is observed, which highlights the exact equivalence of both approaches.

For comparison with these results for the single-atom response under bichromatic driving, we calculate the same quantities – the ladder and crossed double scattering spectra – using the familiar two-atom master equation approach [11, 12, 13]. As already mentioned, this approach provides a general framework for treating intense laser field-atoms interactions including interatomic correlations induced by the exchange of photons, but is restricted to small numbers of scatterers. Both, $I_L(\omega_D)$ and $I_C(\omega_D)$, derived from the two-atom master equation, are found to coincide perfectly with the results of Eqs. (4,5), for all tested sets of the driving field parameters, see Fig. 2. Motivated by this agreement between the two apparently very different approaches, we succeeded to prove their equivalence analytically – at least for the elastic component of the scattered light [17]. We expect that the proof can be extended to the inelastic component, as suggested by the good numerical agreement ob-

served in Fig. 2. Details of the proof will be given elsewhere.

Let us stress again that the reduction of the two-atom problem to single-atom Bloch equations is possible only in the case of large distance $k_L r \gg 1$ (and hence $|g| \ll 1$) between the atoms. In this regime, both atoms exchange only single photons, what, in turn, allows to describe the field acting on each atom by a classical field, see Eq. (1). The same argument remains valid for a multiple scattering process with more than two atoms. Provided that the atoms are far away from each other, there are again only single photons exchanged between the individual atoms. Moreover, the possibility that two photons emitted by the same atom meet again at another atom before leaving the sample can be neglected. In other words, all photons incident on a particular atom originate from different previous atomic scatterers, and are hence uncorrelated with each other. Since the quantum properties of the electromagnetic field manifest themselves only in correlations between different photons, the field inside the disordered sample can therefore be described by a classical field. This allows, in principle, to generalize the diagrammatic multiple scattering theory valid for clas-

sical nonlinear scatterers [8] to the atomic case. The main difficulty to be addressed in future work will be to find an efficient method to solve the *single atom* optical Bloch equation for a fluctuating, polychromatic field, representing the radiation emitted by many randomly placed atoms.

In summary, we have shown that the spectrum of intense laser light scattered by two distant atoms can be reproduced by solving the quantum mechanical time evolution equations for a *single atom* in the presence of a weak probe field representing the light emitted by the other atom. The interaction between the atoms is fully taken into account by integrating over the frequencies of the exchanged photons. Since, in contrast to other standard methods in theoretical quantum optics, it is *not* necessary to perform calculations in the composite Hilbert space of both atoms, our method will exhibit very favourable scaling with increasing system size, and can be applied to a larger number of atoms. Thereby, it can serve as a starting point for the development of a theory of multiple scattering of intense laser light by optically thick samples of cold atoms.

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- [1] G. Labeyrie, F. de Tomasi, J.-C. Bernard, C. A. Müller, C. Miniatura and R. Kaiser, Phys. Rev. Lett. **83**, 5266 (1999).
 - [2] P. W. Anderson, Phys. Rev. **109**, 1492 (1958).
 - [3] H. Cao, Waves Random Media **13**, R1 (2006).
 - [4] M. D. Lukin, Rev. Mod. Phys. **75**, 457 (2003).
 - [5] E. Akkermans, G. Montambaux, J.-L. Pichard, and J. Zinn-Justin (Eds.) *Mesoscopic Quantum Physics* (Elsevier, Amsterdam, 1994).
 - [6] V. M. Datsyuk, I. M. Sokolov, D. V. Kupriyanov, and M. D. Havey, Phys. Rev. A **74**, 043812 (2006).
 - [7] A. Lagendijk and B. A. van Tiggelen, Phys. Rep. **270**, 143 (1996); M. C. W. van Rossum and Th. M. Nieuwenhuizen, Rev. Mod. Phys. **71**, 313 (1999).
 - [8] T. Wellens and B. Grémaud, Phys. Rev. Lett. **100**, 033902 (2008).
 - [9] T. Wellens and B. Grémaud, arXiv 0905.3145 (2009).
 - [10] T. Wellens, B. Grémaud, D. Delande, and C. Miniatura, Phys. Rev. A **70**, 023817 (2004).
 - [11] V. Shatokhin, C. A. Müller, and A. Buchleitner, Phys. Rev. Lett. **94**, 043603 (2005).
 - [12] V. Shatokhin, C. A. Müller, and A. Buchleitner, Phys. Rev. A **73**, 063813 (2006).
 - [13] V. Shatokhin, Opt. Spectrosc. **103** 300 (2007); V. Shatokhin, T. Wellens, B. Grémaud, and A. Buchleitner, Phys. Rev. A **76**, 043832 (2007).
 - [14] B. Grémaud, T. Wellens, D. Delande, and C. Miniatura, Phys. Rev. A **74**, 033808 (2006).
 - [15] C. Cohen-Tannoudji, J. Dupont-Roc, G. Grynberg, *Atom-Photon Interactions* (John Wiley & Sons Inc, New York, 1992).
 - [16] B. R. Mollow, Phys. Rev. **188**, 1969 (1969).
 - [17] T. Geiger, *New approach to multiple scattering of intense laser light by cold atoms*, Diploma thesis, Albert-

Ludwigs-Universität Freiburg (2009).